	A STATE OF THE STA	OMMERCE PATENT AND TRADEMARK OFFICE	ATTORNEY'S DOCKET NUMBER  KUBOMURA 1
		TO THE UNITED STATES	U.S. APPLICATION NO (If known, see 37 CFR 1.5)
		ED OFFICE (DO/EO/US) NG UNDER 35 U.S.C. 371	09/720372
CO	INCERNING A FILL		
	TIONAL APPLICATION NO.	INTERNATIONAL FILING DATE  24 April 2000	PRIORITY CLAIMED 23 April 1999
PCT/JI	P00/02652	Z4 APITI 2000	23 April 1333
FIBER	NVENTION -REINFORCED RESI R EXPANSION	IN COMPOSITE MATERIAL	HAVING REDUCED COEFFICIENT OF
	T(S) FOR DO/EO/US U <b>BOMURA et al.</b>		
1. [X] 2. [ ] 3. [X] 3. [X] 4. [X] 5. [X] 6. [ ] 7. [X] 1. [8. [ ] 1. [9. [ ] 1. [1] 1	This is a <b>FIRST</b> submission. This is a <b>SECOND</b> or <b>SUB</b> . This is an express request to examination until the expirate US has been elected in a A copy of the International A a. [ ] is attached hereto (r b. [X] has been communicated. [ ] is not required, as the An English language translated Amendments to the claims of a. [ ] are transmitted here b. [ ] have been communicated. [X] have not been maded. [X] have not been maded An English language translated An oath or declaration of the An English language translated (35 U.S.C. 371(c)(5)).	and of items concerning a filing under 35 U.S. <b>SEQUENT</b> submission of items concerning a begin national examination procedures (3: ation of the applicable time limit set in 35 and Demand by the expiration of 19 months for application as filed (35 U.S.C. 371(c)(2)) equired only if not transmitted by the Internated by the International Bureau. The application was filed in the United State ation of the International Application as filed if the International Application under PCT are with (required only if not transmitted by the international Bureau.  It is, however, the time limit for making such a set and will not be made. It is to determine the content of the annexes to the International Preference inventor(s) (35 U.S.C. 371(c)(4)) the sum of the annexes to the International Preference inventor(s) (35 U.S.C. 371(c)(4)).	g a filing under 35 U.S.C. 371.  5 U.S.C. 371(f) ) at any time rather than delay U.S.C. 371(b) and PCT Articles 22 and 39(1).  from the priority date (PCT Article 31).  mational Bureau).  s Receiving Office (RO/US).  d (35 U.S.C. 371(c)(2)).  Article 19 (35 U.S.C. 371(c)(3))  te International Bureau).  mendments has NOT expired.
11. [ 12. [ 13. [X] [X 14. [ 15. [	An Information Disclosure An Assignment document for A FIRST preliminary amend A SECOND or SUBSEQUE A substitute specification. A change of power of attorn Other items or information: X Courtesy copy of the Int X Courtesy copy of the fir X Formal drawings, 3 she X Courtesy Copy of the Int	Iment. ENT preliminary amendment.  ey and/or address letter.  ternational Application as filed. est page of the International Publication (Wests, Figures 1-4.	apliance with 37 CFR 3.28 and 3.31 is included.  O 00/64668).

U.S. APPLICA ROWN, see 37 CFR 1	Attorney's Docket N				
09/72037	KUBOMURA	1			
17. [xx] The following fees are submi	CALCULATIONS	PTO USE ONLY			
BASIC NATIONAL FEE (37 CFR 1. Neither international preliminary example)		ED 1 492)			
nor international search fee (37 CFR)	1.445(a)(2)) paid t	USPTO			
and International Search Report not p			\$1000.00		
International preliminary examination USPTO but International Search Rep			5960.00		
OSF 10 out international Search Rep	ort prepared by the	EFO of JPO	3060.00		
International preliminary examination	n fee (37 CFR 1.48	(32) not paid to USP	O but		
international search fee (37 CFR 1.44	15(a)(2)) paid to U	SPTO	\$710.00		
International preliminary examination	a faa maid ta ITCDYI	O /27 CED 1 493)			
but all claims did not satisfy provisio	ns of PCT Article	33(1)-(4)	\$690.00		
out an elamin ara not savinly provint		33(1) (1)			
International preliminary examination					
and all claims satisfied provisions of	PCT Article 33(1)	-(4)	\$100.00		
		A CITAC TOTAL A BATA	OTINE	\$ 860.00	
Surcharge of \$130.00 for furnishing the		ASIC FEE AM			
months from the earliest claimed priori			[ ]30	\$ 130.00	
Claims as Originally Presented	Number Filed	Number Extra	Rate		
Total Claims	6 - 20		X \$18.00	\$	
Independent Claims	2 - 3		X \$80.00	\$	
Multiple Dependent Claims (if applica		L	+\$270.00	\$	
TOT	AL OF ABOV	E CALCULA	TIONS =	\$ 990.00	
Claims After Post Filing Prel. Amend	Number Filed	Number Extra	Rate		
Total Claims	20 - 20		X \$18.00	\$	
Independent Claims	2 - 3		X \$78.00	\$	
		E CALCULAT		\$ 990.00	
Reduction of 1/2 for filing by small entit	y, if applicable. A	pplicant claims sma	ll entity	\$	
status. See 37 CFR 1.27.		OT ID I		Φ.	
Excessions	. 41 TO 1: 1 .		TOTAL =	\$	
Processing fee of \$130.00 for furnishin menths from the earliest claimed priori			]20 [ ]30	\$	
		TAL NATIONA	L. FEE =	\$ 990.00	
Fee for recording the enclosed assignm				\$	
accompanied by an appropriate cover s	heet (37 CFR 3.28	, 3.31). \$40.00 per p	property +	•	
	TO	TAL FEES ENC	LOSED =	\$ 990.00	
				Amount to be:	\$
				refunded	
- [ ] A -11- :- 41				charged	\$
<ul><li>a. [ ] A check in the amount of \$_</li><li>b. [X] Credit Card Payment Form (PT</li></ul>		ver the above fees is		OO OO in attached	
c. [ ] Please charge my Deposit Acc	ount No. <b>02-4035</b>	in the amount of \$	mount of 9 3;	to cover the above fee	s
A duplicate copy of this sheet	is enclosed.	_		-	•
d. [XX] The Commissioner is hereby	authorized to char	ge any additional fee	es which may	be required, or credit ar	ny overpayment
to Deposit Account No. <b>02-40</b>	35. A duplicate co	opy of this sheet is e	nclosed.		
NOTE: Where an appropriate time	limit under 37 Cl	FR 1.494 or 1.495 h	as not heen n	net a netition terrelive	67 CFR 1 1376) or
(b)) must be filed and granted to res				ici, a pelifon of 1011e	37 CFR 1.157(4) 01
				X 1/1 1	eeweer
SEND ALL CORRESPONDENCE TO:				2 0 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1 1	unac
BROWDY AND NEIMARK,	PLIC			SIGNATURE \	<u> </u>
624 NINTH STREET, N.W.,	Sheridan Neimarl	<u> </u>			
WASHINGTON, D.C. 20001	3011E 300			NAME	
TEL: (202) 628-5197				20,520 REGISTRATION NU	MRER
FAX: (202) 737-3528				INCOMINE	IVIPE! \
Date of this submission: <b>December 26</b> Form PTO-1390 (as slightly revised by Browdy a				Page 2	

# IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of: Kenji KUBOMURA et al.	) Art Unit: ) ) )
IA No.: PCT/JP00/02652	)
IA Filed: 24 April 2000	) Washington, D.C.
U.S. App. No.; (Not Yet Assigned)	) ) )
National Filing Date:  (Not Yet Received)	December 26, 2000
For: FIBER-REINFORCED RESIN	) Docket No •KIIBOMIRA

# PRELIMINARY AMENDMENT

Honorable Commissioner for Patents and Trademarks Washington, D.C. 20231

Sir:

Contemporaneous with the filing of this case and prior to calculation of the filing fee, kindly amend as follows:

# IN THE SPECIFICATION

After the title please insert the following paragraph:

-- The present application is the national stage under 35 U.S.C. 371 of PCT/JP00/02652, filed 24 April 2000.--

£, , , ,

# IN THE CLAIMS

Claim 3, line 3, delete "claims 1 or 2", and insert therefor --claim 1--.

Claim 5, line 3, delete "at least one of claims 1 through 4", and insert therefor --claim 1--.

Claim 6, line 4, delete "one of claims 1 through 5", and insert therefor --claim 1--.

If, inadvertently, a proper multiple dependent claim has not been amended to reduce it to single dependency, please amend it to be dependent solely on the first-mentioned claim, or, if that is not possible, please cancel the claim and notify the undersigned.

## REMARKS

The above amendment to the specification is being made to insert reference to the PCT application of which the present case is a U.S. national stage. The above amendments to the claims are being made in order to eliminate multiple dependency and for the purpose of reducing the filing fee. Please enter this amendment prior to calculation of the filing fee in this case.

Favorable consideration and allowance are earnestly solicited.

Respectfully submitted,

BROWDY AND NEIMARK, P.L.L.C.

Attorneys for Applicant

By: 6heridan Neimark

Registration No. 20,520

SN:wrd

Telephone No.: (202) 628-5197 Facsimile No.: (202) 737-3528

# 09/720372 528 Rec'd PCT/PTO **26** DEC 2000

# IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of:
 Kenji KUBOMURA et al.

IA No.: PCT/JP00/02652

IA Filed: 24 April 2000

U.S. App. No.:
 (Not Yet Assigned)

National Filing Date:
 (Not Yet Received)

For: FIBER-REINFORCED RESIN ...

Docket No.:KUBOMURA 1

# SUPPLEMENTAL PRELIMINARY AMENDMENT

Honorable Commissioner for Patents and Trademarks Washington, D.C. 20231

Sir:

Prior to examination upon the merits, kindly amend as follows:

# IN THE CLAIMS

Please add the following claims:

-7. In-plane quasi-isotropic fiber reinforced resin composite material having a reduced coefficient of linear expansion as claimed in claim 2, wherein said coefficient of linear expansion is reduced by combining sheets woven by one

kind or more than reinforced fibers of which a coefficient of linear expansion is controlled by a three dimensional structure of twisting yarn, biaxial textile or triaxial textile.

- 8. In-plane quasi-isotropic fiber reinforced resin composite material having a reduced coefficient of linear expansion selected from material claimed in claim 7.
- 9. In-plane quasi-isotropic fiber reinforced resin composite material having a reduced coefficient of linear expansion selected from material claimed in claim 2.
- 10. In-plane quasi-isotropic fiber reinforced resin composite material having a reduced coefficient of linear expansion selected from material claimed in claim 3.
- 11. In-plane quasi-isotropic fiber reinforced resin composite material having a reduced coefficient of linear expansion selected from material claimed in claim 7.
- 12. Yarn prepreg, one-way prepreg, biaxial textile, triaxial textile, four axial textile or a prepreg formed by these textile used for in-plane quasi-isotropic fiber reinforced resin composite material as clamed in claim 11,

SN

wherein a coefficient of linear expansion of said material is controlled.

- 13. Yarn prepreg, one-way prepreg, biaxial textile, triaxial textile, four axial textile or a prepreg formed by these textile used for in-plane quasi-isotropic fiber reinforced resin composite material as clamed in claim 10, wherein a coefficient of linear expansion of said material is controlled.
- 14. Yarn prepreg, one-way prepreg, biaxial textile, triaxial textile, four axial textile or a prepreg formed by these textile used for in-plane quasi-isotropic fiber reinforced resin composite material as clamed in claim 9, wherein a coefficient of linear expansion of said material is controlled.
- 15. Yarn prepreg, one-way prepreg, biaxial textile, triaxial textile, four axial textile or a prepreg formed by these textile used for in-plane quasi-isotropic fiber reinforced resin composite material as claimed in claim 8, wherein a coefficient of linear expansion of said material is controlled.

Yarn prepreg, one-way prepreg, biaxial textile, triaxial textile, four axial textile or a prepreg formed by these textile used for in-plane quasi-isotropic fiber reinforced resin composite material as clamed in claim 7, wherein a coefficient of linear expansion of said material is controlled.

SN

17. Yarn prepreg, one-way prepreg, biaxial textile, triaxial textile, four axial textile or a prepreg formed by these textile used for in-plane quasi-isotropic fiber reinforced resin composite material as claimed in claim 5, wherein a coefficient of linear expansion of said material is controlled.

SN

Yarn prepreg, one-way prepreg, biaxial textile, triaxial textile, four axial textile or a prepreg formed by these textile used for in-plane quasi-isotropic fiber reinforced resin composite material as clamed in claim 4, wherein a coefficient of linear expansion of said material is controlled.

50

19. Yarn prepreg, one-way prepreg, biaxial textile, triaxial textile, four axial textile or a prepreg formed by these textile used for in-plane quasi-isotropic fiber

reinforced resin composite material as clamed in claim 3, wherein a coefficient of linear expansion of said material is controlled.

20. Yarn prepreg, one-way prepreg, biaxial textile, triaxial textile, four axial textile or a prepreg formed by these textile used for in-plane quasi-isotropic fiber reinforced resin composite material as claimed in claim 2, wherein a coefficient of linear expansion of said material is controlled.

# REMARKS

Claims 1-20 presently appear in this case. The above amendments to the claims are being made in order to add new claims and to restore at least partly the varying scope of claims which was eliminated by the elimination of multiple dependencies in the claims.

# Favorable consideration is earnestly solicited.

Respectfully submitted,

BROWDY AND NEIMARK, P.L.L.C.

Attorneys for Applicant

By: Sheridan Neimark

Registration No. 20,520

SN:wrd

Telephone No.: (202) 628-5197 Facsimile No.: (202) 737-3528

7

## SPECIFICATION

# FIBER REINFORCED RESIN COMPOSITE MATERIAL HAVING REDUCED COEFFICIENT OF LINEAR EXPANSION

# Field of the Invention

The present invention relates to fiber reinforced resin material having a reduced coefficient of linear expansion, particular relates to fiber reinforced resin material having a coefficient of linear expansion of substantially zero.

# Background of the Invention

In a conventional art, fiber reinforced resin composite material having a reduced coefficient of linear expansion can accomplish physical and chemical characteristics suitable for its purpose by selecting a combination of reinforced fiber and resin matrix, and has been broadly used. Particularly, fiber reinforced resin composite material is strong with respect to chemical reaction and corrosion environment, and has a low specific gravity and high stiffness in general. The material has been broadly utilized as structures and parts in the field of an aircraft and space environment, applicable to parts of fine machinery devices, material of construction or sporting goods.

In the case of utilizing material as structural parts for fine tools used in an environment thermally influenced, it would become a serious problem caused by thermal expansion in accordance with a temperature varying. For example, in the case of an artificial satellite, a temperature at a side confronting with sun and a temperature at a side opposite to sun are remarkably different so that installed fine tools are baneful influenced

Although the second of the sec

by thermal expansion in such an environment. In a field of a semiconductor manufacturing device, an optical tool and a fine processing device, baneful influence caused by thermal expansion should be reduced as less as possible.

In a conventional art, although material having reduced coefficient of thermal expansion such as Inver alloy, quartz glass, carbon fiber reinforced resin composite material has been used, these are metal, silica system material or limited material. the case of the material, its utility, strength, apecific gravity and manufacturing cost would be restricted. It is required a coefficient of liner expansion as less as possible so as to utilize a fiber reinforced resin composite material in the various field described above. In order to control a coefficient of linear expansion of fiber reinforced resin composite material, it is prefer to use reinforced fiber and matrix resin of which a coefficient of linear expansion is low or substantially zero. However, it is not an actual way to provide the characteristic described above with respect to the both of the reinforced fiber and the resin matrix. There has been an experiment to reduce a total coefficient of linearly expansion by combining fiber having a negative coefficient of linear expansion along a fiber direction and fiber having a positive coefficient of linear expansion along the fiber direction so as to cancel these coefficients each other.

However, in the method, it would be difficult for the both of reinforced fiber and resin matrix to provide suitable characteristics. Upon considering strength and heat-resistant of the fiber reinforced resin composite material originally required, a material selection is so restricted.

A factor for determining a coefficient of linear expansion

of a plate member and a furnished product made of reinforced fiber resin composite material is not only a coefficient of linear expansion of these materials but also a blending ratio of the reinforced fiber and the matrix resin. Depending on a kind of reinforced fiber, such as monofilament, yarn doubling and yarn twisting, a structure of prepreg and a laminating condition such as a three dimensional laminating structure formed by biaxial/triaxial textile with one-way prepreg aligned along one direction and a structure of filaments and their laminated structure.

The present inventors reviewed a combination of reinforced fibers and their kinds, a selected structure of the reinforced fiber and a prepreg so as to provide reinforced fiber resin composite material having superior characteristics wherein the inventors found factors how a coefficient of linear expansion can be effectively and finely reduced.

# Disclosure of the Invention

T.

IJ

Regarding an in-plane quasi-isotropic material formed by reinforced fiber resin composite material according to the present invention, two kinds or more than reinforced fibers are combined, wherein at least one reinforced fiber having a negative coefficient of linear expansion is included. A respective sheet woven by one kind or more than reinforced fibers of which each coefficient of linear expansion is controlled is combined so as to reduce a total coefficient of linear expansion, wherein two kinds or more than reinforced fibers are formed as a strand in a condition of monofilament, yern doubling or twisting yarn.

Under the above structure, a plurality of sheets formed by one kind or more than reinforced fiber in which a three

dimensional structure of twisting yarn, biaxial textile or triaxial textile controls its coefficient of linear expansion are combined so as to provide an in-plane quasi-isotropic material by providing reinforced fiber resin composite material wherein the coefficient of linear expansion is reduced.

Further, two or more than sheets made of reinforced fiber are combined wherein a coefficient of linear expansion of the respective reinforced fiber is different each other and at least one reinforced fiber has a negative coefficient of linear expansion so as to provide an in-plane quasi-isotropic material by providing reinforced fiber resin composite material wherein the coefficient of linear expansion is reduced. These kinds of reinforced fibers can be selected from various selections. Even if a reinforced fiber had an excellent elastic modulus and excellent chemical characteristics, the reinforced fiber could not have been utilized since the reinforced fiber has a negative coefficient of linear expansion. Unless a reinforced fiber had a proper coefficient of linear expansion, the reinforced fiber could not have been utilized. Nowadays, these reinforced fibers can be utilized in the present invention.

By combining these reinforced fibers in a form of one prepreg or a plurality of prepreg, a total coefficient of linear expansion is controlled to a predetermined level. After forming reinforced fiber resin composite material with these reinforced fibers, a final coefficient of the material would be within a preferable range, for example, substantially zero. Regarding the combined reinforced fibers, the respective coefficient of linear expansion of monofilament is not so important. The coefficient of linear expansion of the reinforced fiber with respect to a resin matrix and the coefficient of the resin matrix are finally controlled

والرابي والمراوي والمهاروم وميروم ومناه والمنافي والمنافية والمنافية

so that a reduced coefficient of linear expansion can be provided. Upon considering a structure of reinforced fiber formed by monofilament, a coefficient of linear expansion of the combined reinforced fibers is controlled by selecting kinds and number of reinforced fibers and its blending ratio.

Embodiments of these reinforced fibers may be yarn doubling of which the total coefficient of linear expansion is controlled or a strand formed by two kinds or more than reinforced fibers are bundled.

In the present invention as claimed in claim 3, a coefficient of linear expansion is controlled by utilizing a fact that the coefficient is varied by a three dimensional structure of reinforced fiber. A three dimensional structure is formed by twisting yarn in which reinforced fibers are twisted so that a substantial length becomes longer and a twisting condition is changed caused by expansion/contraction of the twisting yarn. As the result, an apparent coefficient of linear expansion is changed. Thus, a substantial coefficient of linear expansion can be varied by utilizing such a twisting yarn. By preparing a prepreg formed by reinforced fibers made of the twisting yarn, a coefficient of linear expansion of a fiber reinforced structure can be controlled by utilizing the same kind of reinforced fiber. As similarly, by preparing one-way prepreg, prepreg formed by biaxial textile or prepreg formed by triaxial textile, the substantial length of the fiber is different depending on their three dimensional textile structures - Apparent coefficient of linear expansion is varied caused by expansion/contraction of the prepreg. Upon considering the difference of the coefficients of linear expansion caused by the structures of the prepregs, the total coefficient of linear expansion of fiber reinforced

resin composite material can be reduced to a predetermined level.

In the present invention as claimed in claim 4, a coefficient of linear expansion of a laminated reinforced structure is controlled by combining sheets with different coefficients of linear expansion made of reinforced fibers having different coefficients of linear expansion. Regarding these sheets having different linear expansion ratio, a sheet aligned along one direction and a sheet formed by biaxial/triaxial textile may be utilized. A coefficient of linear expansion is controlled by laminating a predetermined number of pairs including two or more than sheets having different coefficients of linear expansion so as to reduce a coefficient of linear expansion of fiber reinforced resin composite material to a predetermined level as an in-plane quasi-isotropic material.

As described above, by combining reinforced fibers having different coefficient of linear expansion, or considering a three dimensional structure of sheets, the coefficient of linear expansion is controlled by laminating sheets formed by reinforced fibers having a different coefficient of linear expansion so that a coefficient of linear expansion of reinforced fiber resin to composite material can be controlled to a predetermined level. In addition, by combining these structures, a total coefficient of liner expansion can be controlled more effectively and easily.

## Brief Description of the Drawings

Fig. 1 is a schematic view for explaining a control of a coefficient of linear expansion of fiber reinforced resin composite material.

Fig. 2 shows a structure bundled by reinforced fibers A and B, wherein each coefficient of linear expansion is different.

And the second services of the second second second services and

Fig. 3 shows an example of a prepreg sheet formed by bundled fibers of which the respective coefficient of linear expansion is different each other. Fig. 3(a) shows a one-way prepreg sheet, Fig. 3(b) shows a biaxial prepreg sheet and Fig. 3(c) shows a triaxial prepreg sheet.

Fig. 4 shows an example of fiber bundle having a constant coefficient of linear expansion formed by reinforced fibers having different coefficient of linear expansion. Fig. 4(a) is an example of the one-way direction prepriety, Fig. 4(b) is an example of the biaxial prepriety sheet; and Fig. 4(c) is an example of the triaxial prepriety sheet.

The Preferred Embodiment of the Present Invention

With reference to the accompanying drawings, it will be explained a concrete structure of fiber reinforced resin composite material according to the present invention and its embodiments.

Fig. 1 shows a principle how a coefficient of linear expansion of an in-plane quasi-isotropic plate according to the present invention is controlled to a predetermined level by combining a sheet 1 and a sheet 2 having different coefficient of linear expansion each other. In Fig. 1, one reinforced fiber has a negative coefficient of linear expansion. Regarding the other reinforced fiber, its coefficient may be relatively large (positive). By alternatively laminating the sheet 1 and the sheet 2 as shown in Fig. 1 in order to provide an in-plate quasi-isotropic plate and thermally hardening the plate, a coefficient of linear expansion formed as fiber reinforced resin composite material can be controlled to zero or a predetermined level.

Under the structure described above, a coefficient of linear expansion of each reinforced fiber and a coefficient of linear

expansion of matrix resin can be selected easily. Under the combination, fiber having a high elastic coefficient and reinforced fiber having excellent chemical characteristics may be blended with reinforced fiber of which material is suitable for controlling a coefficient of linear expansion so as to provide new material having the most preferable characteristics.

Fig. 2 shows a bundle of reinforced fibers formed by at least two kinds of fibers, a reinforced fiber A with one coefficient of linear expansion and a reinforced fiber B with a different coefficient of linear expansion. By blending these two kinds of reinforced fibers, a strand of which a coefficient of linear expansion is controlled to a predetermined value can be obtained.

By bundling reinforced fibers, bundled fibers can be treated as one material having a constant coefficient of linear expansion so that composite material can be formed by a one-way prepreg by aligning these bundles along one direction, a biaxial textile prepreg or a triaxial textile prepreg. Accordingly, by laminating a predetermined number of prepreg formed by fiber bundles having a constant coefficient of linear expansion, a coefficient of linear expansion, a coefficient of linear expansion of an in-plate quasi-isotropic plate can be controlled.

Fig. 3(a) shows a one-way prepred wherein fiber bundles A and B formed by reinforced fibers having different coefficient of linear expansion are aligned along one direction, Fig. 3(b) shows a biaxial textile prepred and Fig. 3(c) shows a triaxial textile prepred. The coefficient of linear expansion can be controlled to the predetermined level by alternatively aligning fiber bundles A and B having different coefficient of linear expansion each other. A product process of the material is as similar as that of normal reinforced fiber resin composite

material.

T.

Ш

Fig. 4(a) through 4(c) shows examples of fiber bundles 3 having a predetermined value of linear expansion by blending reinforced fibers having different coefficient of linear expansion as shown in Fig. 2, respectively. In these examples, the fiber bundles have the same coefficient of linear expansion so that the fiber bundles can be treated as similar as normal fiber bundles. In an example as shown in Fig. 4(a), a diameter of a monofilament is 4 to  $50\,\mu$  m and number of fibers are 10 to 1000,000. By aligning these fibers as shown in Fig. 4(a), one-way prepreg having a thickness of 10 to  $500\mu$  m and a width of 1 to 150cm can be provided.

Fig. 4(b) and 4(c) is examples of biaxial textile and triaxial textile, respectively.

Regarding a combination of reinforced fibers according to the present invention, actual cases of the combination of reinforced fiber and resin matrix are shown as follows:

(1) Characteristics of epoxy system resin used for a matrix Tensile elastic modulus: 360kg/mm<sup>2</sup>

Poisson ratio:

60ppm/°C Although the contract of the contract of Thermal expansion ratio:

(2) Characteristics of Reinforced fiber: fiber type, coefficient of linear expansion and tensile elastic modulus of reinforced fiber are shown in Table 1

Table 1: fiber type, coefficient of linear expansion and tensile elastic modulus of reinforced fiber

And the second of the second o

	e franç	
	2	
-	1111	
Ç,	Mann 2	
	Ž,	
2	de de	
-	Hines	
C	Thurs.	
C	-	
	į	
Ē		
	1	
ï	***	

	Fiber type	Tensile elastic modulus along a fiber direction kgf/mm²	Coefficient of linear expansion along a fiber direction x10 <sup>-6</sup> /°C
	PAN-based carbon fiber (M50J)	48,500	- 1
G-	PAN-based carbon fiber (M50J)	60,000	-1-1
	PAN-based carbon fiber (M50J)	23,500	-0.41
	PAN-based carbon fiber (M50J)	35,000	-0.73
	Pitch-based carbon fiber (YS-35)	35,690	-1.1
	Pitch-based carbon fiber (YS-60)	61,183	-1.5
	Pitch-based carbon fiber (YS-70)	71,972	-1.5
	Aramid fiber (K149)	19,000	<b></b> 2
	PBO fiber (zairon)	28,500	<b>-</b> 6

PBO: Polyparaphenylene Benzo Oxazale

Zairon: trademark registered by TOYO BOSEKI Co., Ltd.

A sheet is formed by arranging these reinforced fibers along one direction and immersed in matrix resin as one-way directional material. In the case of an in-plane quaci-isotropic plate formed by laminating these sheets, a coefficient of linear expansion of the plate is calculated by the following equation (the equation and its explanation were disclosed by one of inventors at a seminar of Composite Material at Hokuriku division on April 14, 1999).

These reinforced fibers are formed as one-way sheet, biaxial textile or triaxial textile. An in-plane quasi-isotropic material is formed by laminating these sheets and hardened so as to form composite material. The coefficient of linear expansion of the respective fiber and sheet in the composite material can be judged in a macro view. Even if the structure of the composite material is different, the total coefficient can be treated usually.

A blend ratio in the case of bundled fibers as shown in

Fig. 2 formed by blanding two kinds of fibers having different coefficient of linear expansion is recognized as a hybrid ratio. A ratio of combining a yarn prepreg, a one-way prepreg, a biaxial propreg, a triaxial prepreg and a four-axial prepreg formed by bundled fibers of two kinds of libers having the different coefficient of linear expansion can be treated as a hybrid ratio.  $\alpha = \left\langle E_L^{+} \alpha_L + E_L^{+} \alpha_T + \mu_{LT} * E_L^{+} \alpha_L + \mu_{LT} * E_L^{+} \left( \alpha_L + \alpha_T \right) \right\rangle / \left( E_L + E_T + 2 \mu_{LT} * E_L \right)$ 

In the equation,

\*lastic modulus of one-way material along a fiber direction E<sub>L</sub>; elastic modulus of one-way material along a direction E-: perpendicular to the fiber direction

 $\mu_{LT}$ : Poisson ratio

Πij

coefficient of linear expansion of one-way material along a fiber direction

 $\alpha_{\text{T}}$ : coefficient of linear expansion of one-way material along a direction perpendicular to the fiber direction

Material characteristics of these one-way materials are determined by kind of fiber and resin and a content ratio of fiber. Regarding characteristics of reinforced fiber, an elastic modulus of one-way material along a transversal direction or a crimp ratio is of textile are influenced. However, there is not sufficient data about them. Therefore, regarding the influence caused by these factors are estimated based on an experimental knowledge and checked actually.

- (3) An in-plane quasi-isotropic plate is formed by laminating sheets made of one-way prepreg and prepreg formed by biaxial textile in accordance with the following method.
- (a) A prepreg is formed by a pair of one-way prepreg formed by a layer 1 and a layer 2 as shown in Fig. 1. These pairs are laminated along directions of 0°,90°, 45°,-45°,-45°,

 $45\,^\circ$  ,  $90\,^\circ$  ,  $0\,^\circ$  in order so as to form an in-plane quasi-isotropic plate.

- (b) Eight sheets of one-way prepregs having the same characteristics as shown in Fig. 4(a) formed by fibers as shown in Fig. 2 are laminated along direction of 0°,90°, 45°, -45°, -45°, 45°, 90°, 0° in order so as to form an in-plane quasi-isotropic plate.
- (c) In the case of biaxial textile, if a crimp ratio is controlled to reduce, the textile is not so different from the one-way material and substantially utilized as same as the one-way prepreg. The similar plate can be produced by laminating sheets of the biaxial textile along a direction of 0°, 45°, 45°, 0°. These four prepregs are same material formed by yarn doubling of two kinds or more than fibers, combing wool or a textile alternatively woven by these fiber bundles. Such a structure is shown in Fig. 2, Fig. 3(b) and Fig. 4(b).
- (d) Regarding an in-plane quasi-isotropic plate formed by the above described method, a relation between a hybrid ratio calculated by the above equation, and a coefficient of thermal expansion is shown in Table 2.

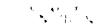
Table 2: Hybrid ratio of fiber and coefficient of thermal expansion of an in-plane quasi-isotropic plate

	_							
G.	Fiber	type			%) = wei veight o			
	Fiber A	Fiber B	0%	20%	40%	60%	80%	100%
	M50J	Meol	-0.21	-0.14	-0.06	0.02	0.11	0.21
-	YS-35	YS-60	-0.39	-0.24	-0.07	0.18	0.41	0.74
	K149	YS-70	-0.55	-0.36	-0.11	0.28	0.93	2.24

(Note)

Lamination direction: 0°,90°, +45°, -45°, -45°, +45°, 90°,  $0^\circ$ 

A CONTROL OF THE PROPERTY OF T



vf= 60%

As shown in Table 2, it would be found a minimum coefficient of thermal expansion depending on the hybrid ratio. Hybrid ratios of reinforced fibers of which a total coefficient of thermal expansion is zero are shown in Table 3.

Table 3: Weight ratio of Fibers A and B, and elastic modulus in the case that an in-plane coefficient of thermal expansion is zero

Fiber ty	pe	Weight of	Weight of fiber (%)			
Fiber A	Fiber B	Fiber A	Fiber B	modulus (kg/mm²)		
M50J	MeOJ	55.1	44.9	11,328		
YS-35	YS-60	44.6	55.4	10,619		
K149	YS-70	45.4	53.6	10,099		

(Note) Vf=60%

- (4) In the case of a triaxial textile, an in-plane quasi-isotropic characteristic can be obtained by one textile sheet. In such a case, a crimp ratio is varied depending on its thickness. Therefore, it has been checked ratios of textile having thickness of 0.1mmm and 0.2mm.
- (a) A relation between a hybrid ratio and a coefficient of thermal expansion of composite material formed by a triaxial textile having a thickness 0.1mm calculated from the above equation

Table 4: Hybrid ratio of fiber and in-plane thermal expansion coefficient (unit:ppm/ ${\mathbb C}$ )

Fiber	type	Hybrid of fib	ratio( er A + v	%). = wej weight c	ght of ; of fiber	fiber A, B) x 1	(weight
Fibe r A	Fiber B	0%	20%	40%	60%	808	100%
T300	Zairon	-2.86	-1.98	-1.04	-0.04	1.04	2.19
K149	Zairon	-2.86	-2.11	-1.26	-0.27	0.89	2.26
M35J	Zairon	-2.86	-1.98	-1.17	-0.03	0.27	0.92

(Note)

thickness of triaxial textile: 0.1mm

Vf = 60%



As similarly, the hybrid ratio is shown in Table 5 in the case that a thermal expansion coefficient of the combined fibers as shown in Fig. 4 is zero.

Table 5: Weight ratio of fibers A and B, and elastic modulus of which an in-plane thermal expansion coefficient is zero.

والروائق والمراوية والمواردة ومهروم وهدان والأرقى المهداء والمواري المماد

Kind of	fiber	Weight of	Weight of fiber (%)		
Fiber A	Fiber B	Fiber A	Fiber B	modulus (kg/mm²)	
T300	Zairon	60.7	39.3	5,729	
K149	Zairon	65	35	5,038	
M35J	Zairon	72.1	27.9	7,221	

(Note) Thickness of triaxial textile: 0.1mm

(b) Table 6 shows a relation between the hybrid ratio and thermal expansion coefficient of composite material made of triaxial textile having a thickness of 0.2mm calculated from the above equation

Table 6: Hybrid ratio of fiber and in-plane thermal expansion coefficient of triaxial textile (unit: ppm/C)

D-	Fiber	typ	е				ght of f of fiber		(weight 100
ļ	Fiber	A	Fiber B	0 %	20%	40%	60%	808	100%
	T300		Zairon	-2.80	-1.94	-1.02	-0.03	1.05	2.22
-	K149		Zairon	-2.80	-2.08	-1.24	-0.26	0.90	2.31
Į	M35J		Zairon	-2.80	-1.95	-1.16	-0.42	0.28	0.95

(Note) a thickness of triaxial textile: 0.2mm,

Vf = 60%

As similarly, the hybrid ratio is shown in Table 7 in the case that a thermal expansion coefficient of the combined fibers as shown in Fig. 6 is zero

Table 7: Weight ratio of Fibers A and B, and elastic modulus in the case that an in-plane quasi-isotropic thermal

# expansion coefficient is zero.

Fiber type		Weight of	Elastic	
Fiber A	Fiber B	fiber A	Fiber B	modulus (kg/mm <sup>2</sup> )
T300	Zairon	60.6	39.4	5,713
K149	Zairon	64.8	35.2	5,028
M35J	Zairon	71.8	28.2	7,168

(Note)

Thickness of triaxial textile: 0.2mm

Vf-60%

In the above embodiments, epoxy resin is used as a matrix. However, if any resins have proper characteristics, various thermoplastic resin can be used as the matrix. In addition, a matrix of composite material including resin and metal may be used if the matrix has proper characteristics.

Further, it is effective to control coefficient of thermal expansion of these resin. Characteristics of resin such as a coefficient of thermal expansion may be controlled by blending various fillers in the resin.

Although the above embodiments show an isotropic coefficient of linear expansion by providing fiber reinforced resin composite material having an in-plane quasi-isotropic characteristic, a combination of fiber bundles and prepreg disclosed in the above embodiments may be controlled in the case that a coefficient of Linear expansion is different depending on its direction-

Industry Field Applicable to the Invention

In fiber reinforced resin composite material according to the present invention, reinforced fiber having various characteristics and different coefficient of linear expansion and matrix resin are combined in the above described embodiments. By controlling a coefficient of linear expansion of reinforced fibers, fiber reinforced resin composite material having a



controlled coefficient of linear expansion and preferable characteristics can be obtained. The material can be broadly utilized as a structure and a part used in a fine machinery or an air craft and space environment where varying temperature and thermal expansion caused by varying temperature are remarkably influenced.

Claims:

- 1. In-plane quasi-isotropic fiber reinforced resin composite material having a reduced coefficient of linear expansion by combining sheets woven by one kind or more than reinforced fibers, each coefficient of linear expansion of said sheets controlled to be reduced by combining two kinds or more than of reinforced fibers wherein said reinforced fibers includes at least one kind of reinforced fibers having a negative coefficient of linear expansion.
- 2. In-plane quasi-isotropic fiber reinforced resin composite a material having a reduced coefficient of linear expansion as claimed in claim 1, said material characterized in that monofilament, yarn doubling or blending strand is used for said two kinds or more than of reinforced fibers.
- In-plane quasi-isotropic fiber reinforced resin composite material having a reduced coefficient of linear expansion as claimed in claims 1 or 2, wherein said coefficient of linear expansion is reduced by combining sheets woven by one kind or more than reinforced fibers of which a coefficient of linear expansion is controlled by a three dimensional structure of twisting yarn, biaxial textile or triaxial textile.
- 4. In-plane quasi-isotropic fiber reinforced resin composite material having a reduced coefficient of linear expansion by combining sheets with different coefficient of linear expansion woven by one kind or more than reinforced fibers, wherein each coefficient of linear expansion of said reinforced fibers is controlled to be reduced by combining two kinds or more than of reinforced fibers wherein at least one kind of said reinforced fibers has a negative coefficient of linear expansion.
  - 5. In-plane quasi-isotropic fiber reinforced resin composite

A PRODUCTION OF THE CONTROL OF THE C

والمحاور فراورا والموفور والمعروضين ومهروم ومعالا والراف فالمومود والموجود والمحافظ

A Principal Common of the Art of the Commission of the Commission

material having a reduced coefficient of linear expansion selected from material claimed in at least one of claims 1 through 4.

6. Yarn prepreg, one-way prepreg, biaxial textile, triaxial textile, four axial textile or a prepreg formed by these textile used for in-plane quasi-isotropic fiber reinforced resin composite material as claimed in one of claims 1 through 5, wherein a coefficient of linear expansion of said material is controlled.

# 

ABSTRACT

In fiber reinforced resin composite material, in order to reduce a coefficient of linear expansion to relatively small, particularly to zero, two kind or more than reinforced fibers are combined, wherein at least one reinforced fiber with a negative coefficient of linear expansion is included, prepreg sheet formed  $_{f c}$  them are laminated so as to provide in-plane quasi-isotropic plate. Thus, a total coefficient of liner expansion of fiber reinforced resin composite material is controlled to zero.

A coefficient of linear expansion of doubling yarn and combing wool combined by two kind or more than reinforced fibers is controlled, a coefficient of linear expansion of prepreg sheet formed by these fiber bundles is controlled or a coefficient of linear expansion of fiber bundles formed by two kinds or more than reinforced fibers is controlled by previously controlling the coefficient of linear expansion of the fiber bundles and considering an influence caused by a three dimensional structure of doubling yarn and textile. As the result, the coefficient of linear expansion of the composite material can be reduced as less as possible.

لما يراس والمرابق والربية والمعار ومعروفة أأن والأناف ومعوف والمدود أوالا فالمعار

1/3

Fig. 1

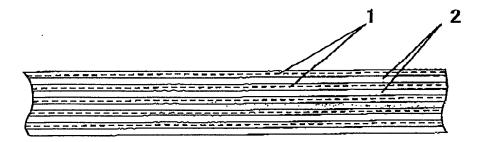
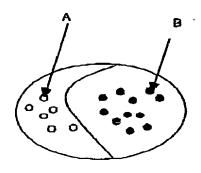


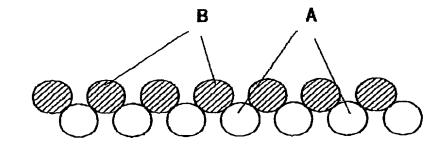
Fig. 2



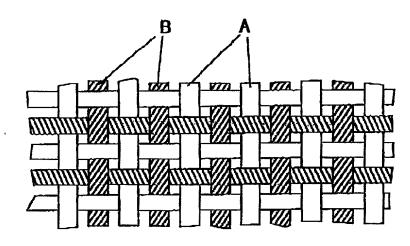
والمرافق والمناجع والموارد ومعروه والمعروض والمراجع والمعروب والمعروب والمعروب والمعروب

Fig. 3



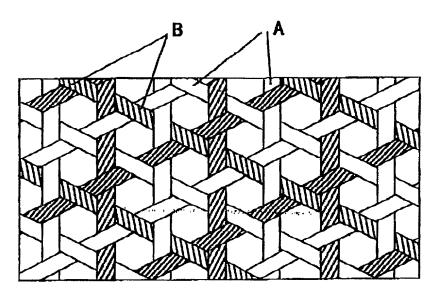


(b)



(c)

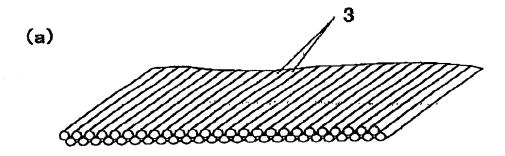
DOVEDSVE . AEDSOI

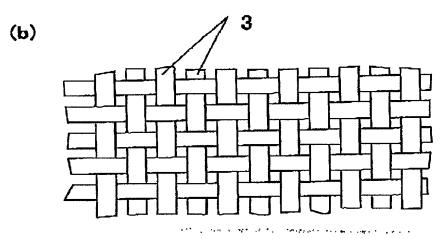


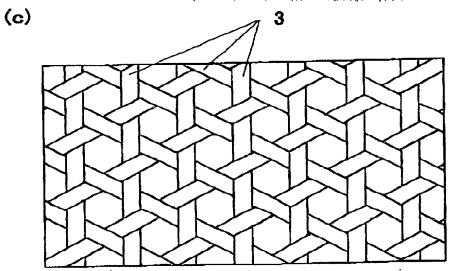
\* \* \* \*

3/3

Fig. 4







The state of the s

Page 1 of 2 Page	s [X]	Original	[ ] Su	ıbstitute	[]	Supplem	ental	Atty. I	Oocket: I	CUBOMUR	A=1
Comb	oined De	claratio	n for P	atent .	Appl	cation	and ]	Power	of Att	orney	
As a below-named in	ventor, I her	eby declare 1	:hat:	-							
My residence, post cand sole inventor (if subject matter which	only one nar	ne is listed b	elow) or	an origina	ıl, first	and joint in	ventor	und that I (if plura)	bolieve I I names ar	am the origi e listed belov	nal, first w) of the
FIBER-REINFORCI			MATER	AL HAV	ING R	EDUCED (	COEFF	ICIENT	OF LINE	AR EXPANS	SION
the specification of v											
[x]		nereto; the United S No. <u>09/72</u>			.C. §11	l on Dece	mber 2	26, 2000	ے as		
[X]		plication,	PCT/ JP0	0/026 <b>52</b> stage app			24	April	, et	ge of an inte itry reques *; §371	
and was amended on		26, 2000 is tes of amends			19 and	34 if PCT)					
I have reviewed and amendment referred known by me to be r	10 above; an	d I acknowl	edge the (	luty to di	sclose 1	o the Pater	on, inc it and 1	eluding th Frademar	ne claims, k Office (	as amended PTO) all infe	l by any ormation
I hereby claim foreig inventor's certificate "Yes" box checked, certificate or PCT in	or §365(a) and have al	of any prior so identified	PCT apple below, by	ication(s)	) design ig the "	ating a cou	intry o	ther than eign appl	the U.S., lication fo	listed below r patent or in	with the
111162	70	J:	ipan			23 April 19	99		(x )	[]	
(Numbe	er)	(Co	untry)			y Month Ye	ır Filed)		YES	NO [ ]	
(Numbe	er)	(Co	untry)		(Da	y Month Yea	ar Filed)	)	YES	NO	
I hereby claim the be	mefit under 3	5 U.S.C. §1	19(e) of a	ny United	States	provis <b>ion</b> al	applic	ations lis	sted below	:	
		(Applica	tion No.)		<b>P</b> -	(Day Monti	Year F	iled)	-		
		(Applica	tion No.)			(Day Monti	Year f	iled)	-		
I hereby claim the b PCT international ap application is not di U.S.C. §112, I ackn C.F.R. §1.56 which date of this application	plication(s) sclosed in so owledge the became avail	designating to the discontinuity to disc	the U.S., I PCT inter lose to th	isted belo national e PTO al	ow and. applicated in a second contraction in a second	insofar as ion in the nation whi	the sul manne ch is m	oject mat r provide naterial te	ter of each and by the patentab	n of the clain first paragra ility as defin	ns of this ph of 32 ned in 37
(Applica	tion No.)		(Day Mo	onth Year	Filed)		(Status	; patentec	, pending,	abandoned)	
(Applica	ation No.)		(Day Mo	onth Year	Filed)	<del></del>	(Status	: patented	, pending,	abandoned)	,
	71 1	* *	C 11					anda whia	1:		

As a named inventor, I hereby appoint the following registered practitioners to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith:

# All of the practitioners associated with Customer Number 001444

Direct all correspondence to the address associated with Customer Number 001444, which is presently:

BROWDY AND NEIMARK, P.L.L.C. 624 Ninth Street, N.W. Washington, D.C. 20001-5303 (202) 628-5197

The undersigned hereby authorizes the U.S. Attorneys or Agents appointed herein to accept and follow instructions from OHTA PATENT OFFICE as to any action to be taken in the U.S. Patent and Trademark Office regarding this application without direct communication between the U.S. Attorneys or Agents and the undersigned. In the event of a change of the persons from whom instructions may be taken, the U.S. Attorneys or Agents appointed herein will be so notified by the undersigned.

П
<b>3</b> 0
in a

2-00

Page 2 of 2 Pages	Atty, Docket: KUBOMURA=1		
Citle: FIBER-REINFORCED RESIN COMPOSITE	Sarial Nia	JUCED COEFF	CIENT
I.S. Application filed PCT Application filed	, Serial-No.		
CT Application filed	, Serial No		
hereby further declare that all statements made herein of and belief are believed to be true; and that these stateme like so made are punishable by fine or imprisonment, or copardize the validity of the application or any patent iss	ents were made with the knowle both, under 18 U.S.C. §1001 and	dec that willful to	use statements and th
PULL NAME OF FIRST INVENTOR	INVENTOR'S SIGNATURE		DATE
KUBOMURA, Kenji	Kongo hubomn	mi \$ 23,2001	
RESIDENCE	70	CITIZENSHIP	
Japan		Japan	
POST OFFICE ADDRESS			
Room 308, Sanpia Yokokawa, 97, Yokokawa 2-ci	home, <u>Kanazawa-shi</u> , Ishikaw	a JPX	
FULL NAME OF SECOND JOINT INVENTOR	INVENTOR'S SIGNATURE		DATE
WATANABE, Akihito	akihito Waturase		Feb. 23. 200/
RESIDENCE		CITIZENSHIP	
Japan		Japan	
POST OFFICE ADDRESS			
14-10 Shimoyasuda, Maruoka-cho, Sakai-gun, Fu	ıkui 910-0363		
FULL NAME OF THIRD JOINT INVENTOR	INVENTOR'S SIGNATURE		DATE
POLL INMAR OF THIRD TORIN MADE AND ADDRESS OF THE POLICE O			
RESIDENCE		CITIZENSHIP	
RESIDENCE .			
POST OFFICE ADDRESS			
FULL NAME OF FOURTH JOINT INVENTOR	INVENTOR'S SIGNATURE	h	DATE
RESIDENCE		CITIZENSHIP	
POST OFFICE ADDRESS		_L	
POST OFFICE ADDRESS			
FULL NAME OF FIFTH JOINT INVENTOR	INVENTOR'S SIGNATURE		DATE
RESIDENCE		CITIZENSHIP	
POST OFFICE ADDRESS			
THE STATE OF OTHER TOP WE WENT TOP	INVENTOR'S SIGNATURE		DATE
FULL NAME OF SIXTH JOINT INVENTOR	11/12/11/2017		
RESIDENCE		CITIZENSHIP	
POST OFFICE ADDRESS			
FULL NAME OF SEVENTH JOINT INVENTOR	INVENTOR'S SIGNATURE		DATE
RESIDENCE		CITIZENSHIP	
POST OFFICE ADDRESS			

ALL INVENTORS MUST REVIEW APPLICATION AND DECLARATION DEFORE SIGNING. ALL ALTERATIONS MUST BE INITIALED AND DATED BY ALL INVENTORS PRIOR TO EXECUTION, NO ALTERATIONS CAN BE MADE AFTER THE DECLARATION IS SIGNED. ALL PAGES OF DECLARATION MUST BE SEEN BY ALL INVENTORS,